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Short communication

Fractal geometry model for through-plane liquid water permeability of fibrous porous carbon cloth gas diffusion layers

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HIGHLIGHTS

- Fibrous porous structure effects on liquid flow in fiber bed are studied.
- A fractal model for through-plane permeability of GDL is proposed based on the fractal analysis method.
- The model is more accurate for the prediction of the through-plane permeability of GDL.

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ABSTRACT

The short communication presents an improved fractal model to predict through-plane liquid water permeability of the gas diffusion layer (GDL) in proton exchange membrane fuel cells (PEMFCs). The porous structure of a GDL is modeled as a combination of parallel and perpendicular channels to the fluid flow direction. The through-plane permeability equation is derived by a hybrid of fractal longitudinal permeability and transverse permeability in parallel with the former. This model is validated by comparing with the referenced data. It is found that the proposed fractal model is preferred for through-plane permeability predictions of GDL.

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1. Introduction

The proton exchange membrane fuel cells (PEMFCs) have shown immense potential as one of the best alternative power source for automotive, stationary and portable applications because of their low emission and relatively high efficiency. The gas diffusion layer (GDL) which is sandwiched between the catalyst layer (CL) and the gas flow channels is a crucial component in the PEMFCs [1]. In general, carbon cloth consisting of randomly packed carbon fibers is commonly used as the GDL. As a transport property, the permeability of the GDL is a key parameter on the performance of the PEMFCs since GDL allows transport of reactant gas, removal of liquid water and conducting electrons. Therefore, numerous research papers aim at predicting the gas permeability of GDL in the past few years. Tamayol et al. [2] theoretically and experimentally investigated effects of mechanical compression and PTFE content on the through-plane gas permeability of GDL of PEM fuel cells. Hao Liang and Cheng [3] used the multiple-relaxation-time (MRT) lattice Boltzmann method (LBM) with multi-reflection solid boundary conditions to study anisotropic permeabilities of a carbon paper gas diffusion layer (GDL). Didari et al. [4] used a geometric modeling scheme called periodic surface model to construct 3D models of GDL microstructure. The through-plane and in-plane permeability of the GDL were computed by solving the incompressible flow field in the reconstructed zone.

The previous experimental results have shown that the microstructures and pore-size distribution of the GDL in PEMFCs have fractal characteristics [5,6]. This means that the fractal theory may be used to predict the gas or water permeability of GDL. Shi et al. [7] presented a fractal permeability model for the GDL. The actual microstructures of the GDL in terms of pore area dimension and tortuosity dimension are accounted in the model. He et al. [8] developed a fractal to predict the permeability and liquid water relative permeability of the GDL (Tornay TGP H-120 carbon paper) in PEMFCs for the hydrophilicity case and the hydrophobicity case. Their research results indicated that a hydrophobic carbon paper is

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preferred for efficient removal of liquid water from the cathode of PEMFCs. Lo et al. [9] investigated the gas permeability for the microporous layer (MPL) and GDL of PEMFCs using fractal method respectively. The gas molecule effect is also considered by using the Adzumi equation. All the above-mentioned fractal models were based on the assumption that the pores within GDL were only considered as a bundle of tortuous capillary tubes with variable cross-sectional area in the through-plane direction. However, randomly oriented, straight, cylindrical fibers of constant diameter are randomly distributed in a unit cell with free overlapping in the carbon paper. The complex structure of pore-level geometry yields numerous capillary/channels not only parallel to fluid flow but also normal to fluid flow in the media. So, the geometrical structure shown in Fig. 1 can be assumed as an idealized representation of the GDL. All gas channels consist of ones which are parallel to the direction of fluid flow (Part A) and ones which are perpendicular to the direction of fluid flow (Part B) (Fig. 2).

In this short communication, we propose a suitable fractal longitudinal and transverse model for predicting the through-plane permeability of fibrous GDL based on the dual-scale porous microstructural characteristics of such fibrous material. Both the parallel and perpendicular channels are included in the fractal model. Some conclusions are to be drawn from the comparisons among proposed fractal model, other existing analytical models and experimental value.

2. Fractal longitudinal permeability K_L

The GDL is usually made of carbon paper which is composed of randomly packed carbon fiber. Porous Part A can be considered as a bundle of tortuous capillary tubes with variable radius for the two-dimensional case. Let the diameter of a capillary in the Part A be λ, and its tortuous length along the flow direction be $L(\lambda)$. Then, these tortuous capillaries can be described by fractal scale equation [10]

$$L(\lambda) = L_0\left(\frac{L_0}{\lambda}\right)^{d_t-1} \tag{1}$$

where L_0 is the characteristic length of a straight channel and d_t the tortuosity fractal dimension with the value of $1 < d_t < 2$. A lower value of d_t corresponds to a lowly tortuous capillary, thus $d_t = 1$represents a straight capillary and $d_t = 2$corresponds to a highly tortuous line that fills whole space. Another characteristic of the fractal object is that the cumulative size distribution of passages/channels follows the power law relation [10]

$$N(L \geq \lambda) = \left(\frac{\lambda_{\max}}{\lambda}\right)^{d_f} \tag{2}$$

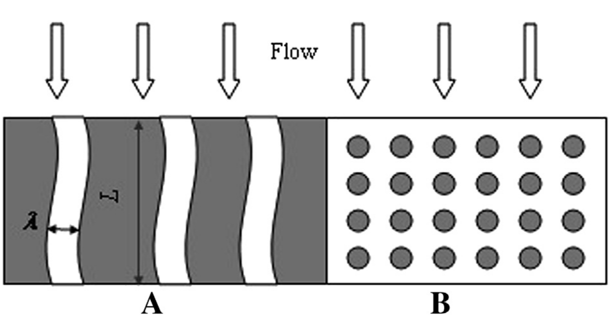

Fig. 1. Porous geometrical structures for GDL.

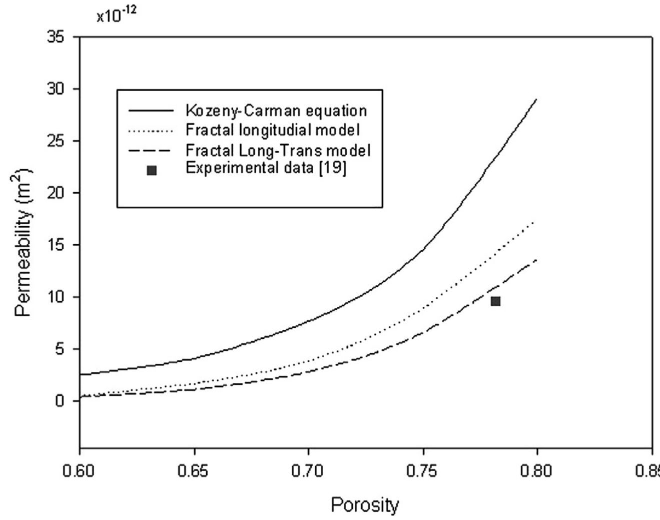

Fig. 2. Comparison between proposed model, referenced data, and other existing models (Long-Trans – longitudinal and transverse).

and

$$-\mathrm{d}N = d_f \lambda_{\max}^{d_f} \lambda^{-(1+d_f)} \mathrm{d}\lambda \tag{3}$$

where d_f is the pore area fractal dimension, λ and $\lambda_{\max}$ are the pore/channel diameter and the maximum pore/channel diameter, respectively.

According to the well-known Hagen–Poiseulle law, one can obtain the flow rate $q(\lambda)$ through a single tortuous capillary. It should be noted that only liquid water permeation is considered, so the Knudsen gas permeability diffusion will not included in the short communication. The $q(\lambda)$ can be given by Ref. [11]

$$q(\lambda) = \frac{\pi}{128}\frac{\Delta p}{L(\lambda)}\frac{\lambda^4}{\mu} \tag{4}$$

where Δp is the pressure gradient, μ is the viscosity of permeation fluid. Then, the total flow rate Q can be calculated by integrating the individual flow rate, $q(\lambda)$ over the entire range of pore sizes from the maximum pore $\lambda_{\min}$ to the maximum pore $\lambda_{\max}$ in a unit cell.

$$Q = -\int_{\lambda_{\min}}^{\lambda_{\max}} q(\lambda)\mathrm{d}N(\lambda) \tag{5}$$

Therefore, the longitudinal permeability K_L of Part A can be written following the Darcy's law [11,21]

$$K_L = \frac{\mu L_0 Q}{\Delta p \cdot A} = \frac{\pi}{128}\lambda_{\max}^{3+d_t}\frac{L_0^{1-d_t}}{A}\frac{d_f}{3+d_t-d_f} \tag{6}$$

where A is the total area of the unit cell and is related to the total pore area A_p by the definition

$$A = \frac{A_p}{\varphi} = \frac{\pi d_f}{4\varphi\left(2-d_f\right)}\lambda_{\max}^2[1-\varphi] \tag{7}$$

Substituting Eq. (7) into Eq. (6) leads to

$$K_L = \frac{1}{32}\lambda_{\max}^{1+d_t}\frac{\varphi}{1-\varphi}L_0^{1-d_t}\frac{2-d_f}{3+d_t-d_f} \tag{8}$$

where φ is the porosity of the GDL. Eq. (8) indicates that the fractal longitudinal permeability is a function of the fractal dimensions for pore area and for tortuosity of tortuous capillaries, porosity, temperature and maximum pore sizes.

3. Transverse permeability K_T

As mentioned previously, numerous perpendicular passages besides parallel channels exist in the fibrous structures. Therefore, the effect of this perpendicular passage orientation to fluid flow on the through-plane permeability of GDL should be considered. Here, the impact can be characterized by transverse permeability. Investigations of the transverse permeability of fibrous porous materials have been widely studied in the past. Sangani and Yao [12] developed an analytical solution for the transverse permeability of randomly packed fiber arrangements up to a fiber volume fraction of 0.7. By using lubrication theory, Bruschke and Advani [13] predicted transverse permeability values close to the fiber packing limit and a simplified cell model to predict the transverse permeability at low fiber volume fractions. Shou et al. [14] studied theoretically transverse flow through aligned fibrous yarns with two length scales and employed Darcy's law and Stokes equation to describe flow behaviors inside the porous yarns and in the open channels between yarns.

It is assumed that the unidirectionally aligned cylindrical fibers are in the rectangular arrangement, as is shown in Fig. 1 (Part B). Tamayol and Bahrami [15] have analytically determined the permeability of ordered arrangements of fibrous media toward normal flow. Especially, analytical relationships are deduced for pressure drop and permeability of rectangular arrangements

$$K_T = d^2 \cdot \left\{ \frac{18\sqrt{\varphi'}\left[\frac{\pi}{2} + \tan^{-1}\left(\frac{1}{\sqrt{\varphi'-1}}\right)\right]}{(\varphi'-1)^{5/2}} + \frac{12\left(\sqrt{\varphi'}-1\right)}{\varphi'\sqrt{\varphi'}}\left[\frac{2-g(\varphi)}{2}\right] + \frac{18+12(\varphi'-1)}{\sqrt{\varphi'}(1-\varphi')^2} \right\} \tag{9}$$

where $\varphi' = \pi/4(1-\varphi)$ and $g(\varphi) = 1.274\varphi - 0.274$. d is the fiber diameter.

4. Total permeability K

In the fibrous media, the permeation is a hybrid of the normal fluid flow and longitudinal fluid flow to aligned fibers. The total permeation resistance through the GDL is that normal flow resistance combined with longitudinal flow resistance in parallel. The total permeability K can be expressed as [15–17]

$$\frac{1}{K} = \frac{\psi}{1-\varphi}\frac{1}{K_T} + \frac{1-\psi}{1-\varphi}\frac{1}{K_L} \tag{10}$$

where ψ is the solid volume fraction of carbon fiber parallel to fluid flow direction. In the considered geometry. Eq. (10) is an analytical solution model of total permeability for GDL. We can substitute Eqs. (8) and (9) into Eq. (10), and get the total gas permeability with the packing geometry parameters in GDL.

5. Results and discussion

Three Toray TGP carbon cloth samples (without PTFE loading) are used in the paper. The pore area fractal dimension and tortuosity fractal dimension can be determined by the box-counting method [18] by processing the SEM image analysis of a unit cell along a plane normal to the principle direction of fluid flow. The fractal dimension values are given in Table 1.

Table 1
Parameters of GDL samples from theory and experimental results.

GDL	d_f	d_t	Referenced permeability (10^{-12} m^{-2})	Fractal Long-Trans model (10^{-12} m^{-2})	Fractal long model (10^{-12} m^{-2})	KC equation (10^{-12} m^{-2})
TGP H-060	1.955	1.157	8.00	9.46	12.18	19.21
TGP H-090	1.891	1.252	8.99	10.02	13.65	22.59
TGP H-120	1.903	1.214	8.69	9.95	12.97	21.65

The permeability of TGP carbon cloth can be obtained by using Eq. (8) integrated with the values of K_L and K_T. The predicted permeability values according to the present fractal Longitudinal-Transverse model are shown in Table 1. For comparison, permeability predictions using the well-known Kozeny–Carman (KC) equation and the fractal longitudinal model analyzed in Section 2 are also shown in Table 1. The semi-empirical KC formula is widely used in the field of flow in porous media and is the starting point for many other permeability models, given by

$$K = \frac{\varphi^{n+1}}{C(1-\varphi)^n} \tag{11}$$

where the exponential n and constant C are called Kozeny–Carman constant parameters. The referenced value of measured permeability is 8.99×10^{-12} m^2 and 8.69×10^{-12} m^2 for TGP H-090 [19] and TGP H-120 [20]. A better agreement was found between permeability results from the fractal Long-Trans model and the referenced data than that from the other two models.

The effects of the porosity of the GDL on the permeability are plotted in Fig. 1 for TGP H-090 carbon cloth. The results of the present fractal model fit the experimental data well. The Kozeny–Carman formula and fractal longitudinal model give higher prediction than predicted permeability by the presented model (fractal Long-Trans model). This reason is that porous materials display higher longitudinal permeability than transverse permeability. In previous fractal models, the fibrous porous medium of GDL is assumed as a bundle of tortuous capillary tubes and only parallel pores to fluid flow direction are taken into account. That is to say, only longitudinal permeability was investigated in these models. In general, KC equation can be used to predict the permeability of such porous media consisting of particles with uniform size distribution. Moreover, the fractal longitudinal model is more appropriate for prediction of upper bound for the permeability and in-plane permeability for fibrous porous bed. The results verify that the fractal Long-Trans model works well on the through-plane permeability predictions of GDL.

6. Conclusions

In this study, a fractal model is proposed to predict the through-plane permeability of the GDL in PEMFCs. The effects of passages orientations including parallel and perpendicular to fluid flow direction on the permeability of fibrous structures are considered. The current fractal model prediction agrees better with the referenced value than that of other analytical models. The fractal Long-Trans model has advantages of more suitable for the prediction of through-plane permeability of fibrous porous media such as GDL. Therefore, fractal geometrical theories serve well as a new research

approach for simulating liquid water transfer through fibrous GDL in PEMFCs.

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